

*Contract no.: FA2386-10-1-4113*

Final report for the project on:

“Transparent Ceramic Yb<sup>3+</sup>:Lu<sub>2</sub>O<sub>3</sub> Materials”

Submission Date: Jan 19<sup>th</sup>, 2012

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Funding level: \$59,000

Grant period: Sep. 24 2010 to Sep. 23 2011

Significant publications: None

Report Documentation Page			Form Approved OMB No. 0704-0188		
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1. REPORT DATE <b>03 FEB 2011</b>		2. REPORT TYPE <b>Final</b>		3. DATES COVERED <b>24-08-2010 to 23-09-2011</b>	
4. TITLE AND SUBTITLE <b>Transparent Ceramic Yb3+:Lu2O3 Materials</b>			5a. CONTRACT NUMBER <b>FA23861014113</b>		
			5b. GRANT NUMBER		
			5c. PROGRAM ELEMENT NUMBER		
6. AUTHOR(S) <b>Akio Ikesue</b>			5d. PROJECT NUMBER		
			5e. TASK NUMBER		
			5f. WORK UNIT NUMBER		
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) <b>World Labo Co., Ltd.,1-2-19-CSJ308 , Mutsuno, Atsuta-ku,Nagoya 456-0023,Japan,NA,NA</b>			8. PERFORMING ORGANIZATION REPORT NUMBER <b>N/A</b>		
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES) <b>AOARD, UNIT 45002, APO, AP, 96338-5002</b>			10. SPONSOR/MONITOR'S ACRONYM(S) <b>AOARD</b>		
			11. SPONSOR/MONITOR'S REPORT NUMBER(S) <b>AOARD-104113</b>		
12. DISTRIBUTION/AVAILABILITY STATEMENT <b>Approved for public release; distribution unlimited</b>					
13. SUPPLEMENTARY NOTES					
14. ABSTRACT <b>Conventional ceramic processing uses a liquid phase co-precipitation method to produce transparent Yb-doped Lu2O3 materials. In this work, fully transparent Yb-doped Lu2O3 ceramics were produced by a simple solid-state reaction method for the first time. The process has very good reproducibility (&gt;60%) based on optical transmittance and interferometry analyses. Optical homogeneity was found to be very good as confirmed by interferometry. Although laser oscillation was achieved with these ceramic specimens, it stopped at 7-8 W input pumping power as sample damage was observed. Further investigation is necessary to determine whether damage was caused by the thickness of the samples due to insufficient heat dissipation or to insufficient optical quality of the developed materials.</b>					
15. SUBJECT TERMS					
16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT <b>Same as Report (SAR)</b>	18. NUMBER OF PAGES <b>15</b>	19a. NAME OF RESPONSIBLE PERSON
a. REPORT <b>unclassified</b>	b. ABSTRACT <b>unclassified</b>	c. THIS PAGE <b>unclassified</b>			

## **Abstract**

Conventionally, liquid phase method (co-precipitation process) is applied to produce transparent Yb doped  $\text{Lu}_2\text{O}_3$  ceramics. In this work, it was successful to produce fully transparent Yb doped  $\text{Lu}_2\text{O}_3$  ceramics by simple solid-state reaction method for the first time. The optical homogeneity of the produced samples was found to be very good as confirmed by interferometry. Laser oscillation was achieved by using the Yb doped  $\text{Lu}_2\text{O}_3$  ceramics. However, laser oscillation stopped at a certain pumping powder level, and damage to the samples was occurred. Further investigations are necessary to find out whether the problem was caused by the thickness of the samples due to insufficient heat dissipation or insufficient optical quality of the developed materials.

## **Background**

After the demonstration of highly-efficient polycrystalline Nd:YAG ceramic laser in 1995, research and development on polycrystalline ceramics for laser application has become worldwide. In recent years, sesquioxide (rare-earth oxides) materials have been paid attention as laser materials because they are very difficult to produce by melt-growth process (conventional single crystal technology). Among them, interesting properties of those materials which are promising for laser application have been reported. However, the optical qualities of the reported materials in the literatures are not sufficient. High laser oscillation efficiency and laser beam quality have not yet been realized, and possibility for power scaling is also at very low condition.

## **Objective of this Program**

Specific gravity of  $\text{Lu}_2\text{O}_3$  is high, and its thermal conductivity is also high. Even it is doped with laser active ions (e.g., Yb or Nd etc.), its thermal conductivity almost does not change. Ionic radius of  $\text{Yb}^{3+}$  ion is very close to that of  $\text{Lu}^{3+}$  ion, and if it is possible to homogeneously dope  $\text{Yb}^{3+}$  into  $\text{Lu}_2\text{O}_3$  host materials, a new type of ceramic laser with the following advantages can be achieved; (1) high quantum efficiency (theoretical quantum efficiency: 91%), (2) almost no concentration quenching, (3) highly thermal conductive gain medium, and (4) possible for power scaling.

Objectives of this program are summarized as follows;

- (1) Selecting the raw materials which is suitable for solid-state method,
- (2) Production of high quality Yb: $\text{Lu}_2\text{O}_3$  ceramics,
- (3) Characterization of optical properties and laser performance of the produced samples, and
- (4) Finding problem points and troubleshooting.

## Summary of Periodical Achievements in the Project

### 1<sup>st</sup> Quarter (2010 Oct-Nov-Dec)

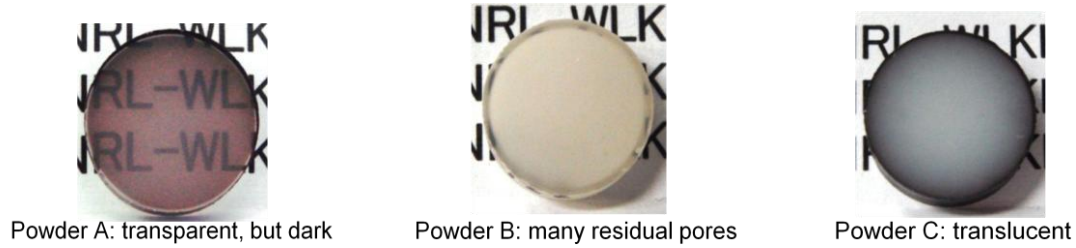


Fig.1a Appearances of the samples produced from different powder sources.

In the 1<sup>st</sup> quarter, screening test for suitable raw powder materials to make test samples to check their sinterability. As shown in Fig.1a, only the power source “A” showed a good transparency, and hence it was selected for the development of this work.

### 2<sup>nd</sup> and 3<sup>rd</sup> Quarter (2011 Jan-Feb-Mar / Apr-May-Jun)

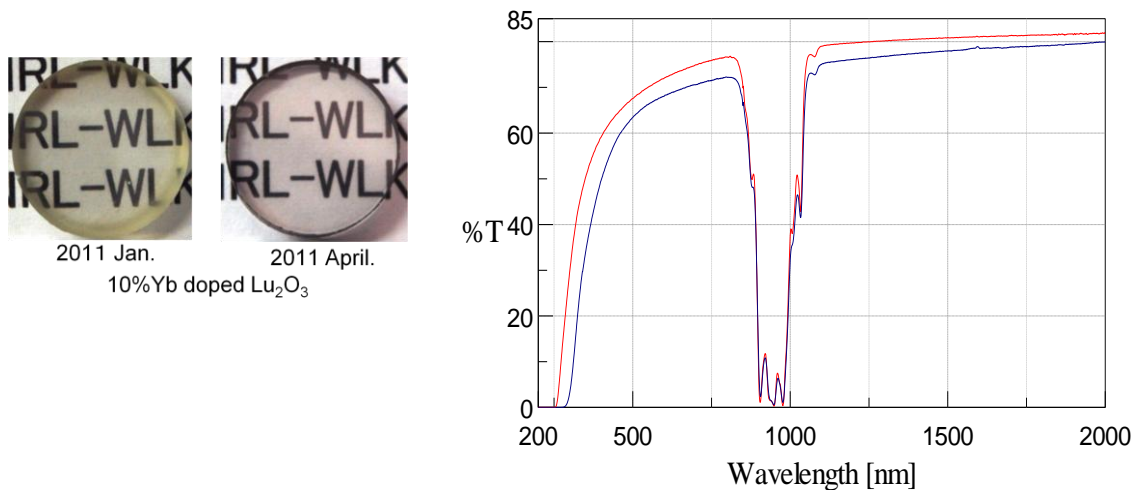


Fig.1b (left) Appearances of the samples produced in 2011 Jan. and 2011 April by using powder “A” source. (right) Comparison of transmittance of these two samples.

In the 2<sup>nd</sup> and 3<sup>rd</sup> quarter, by using sintering additives to improve sinterability, transparent  $\text{Lu}_2\text{O}_3$  ceramics were obtained from 2011 Jan. Also, by improving the milling process condition to homogenize chemically, transparency was improved in 2011 April (please see Fig.1b left). The in-line transmittances of these two samples are compared in Fig.1b (right). As seen in the figure, the improvement of their optical quality was recognized in the whole wavelength range from 200 to 2000 nm.

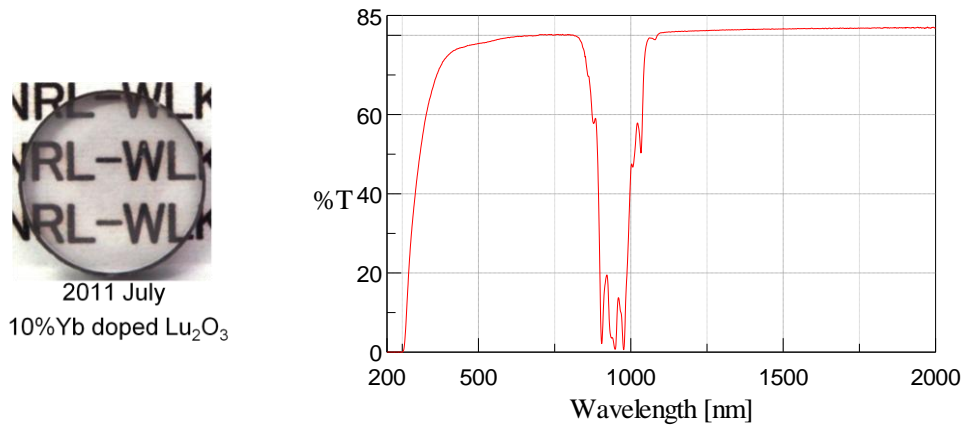


Fig.1c (left) Appearances of the sample produced in 2011 July.  
(right) Transmittance curve of this sample.

In the 4<sup>th</sup> quarter, optimization on the fabrication was performed and reproducibility was also confirmed. As seen the Fig.1c, it was confirmed that the optical quality was further improved.

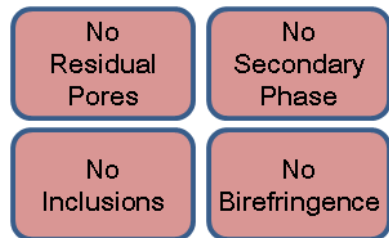


Fig.1d Requirements of target materials for use as laser gain medium

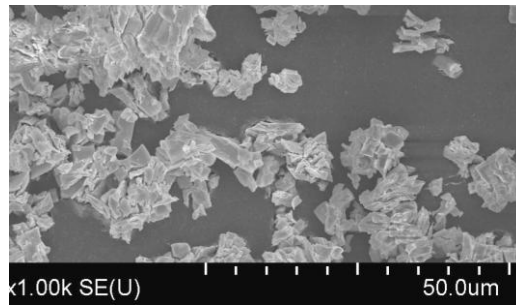
The materials for use as laser gain medium must not include; (1) residual pores, (2) secondary phases, (3) inclusions, (4) birefringence, because they cause scattering and/or absorption problems in the gain medium in laser application. Therefore, if we can achieve material with “no defects & optically homogeneous ceramics” in this work, it will be an excellent laser gain medium.

## Results and Discussion

### Screening Raw Powder materials

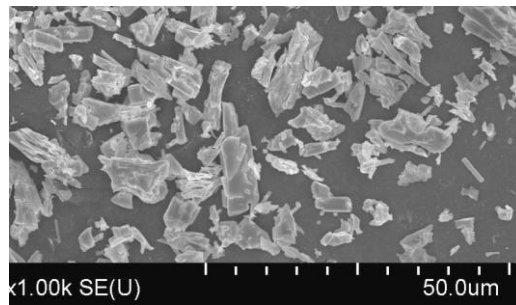
In this work, three different sources of raw powder materials were tested to make transparent ceramics. As seen in Fig.2a, powder “A” has small agglomerations with fine particle shape. Powder “B” has small agglomerations with coarse particle shape. Powder “C” has large agglomerations with slender particle shape.

Powder “A”



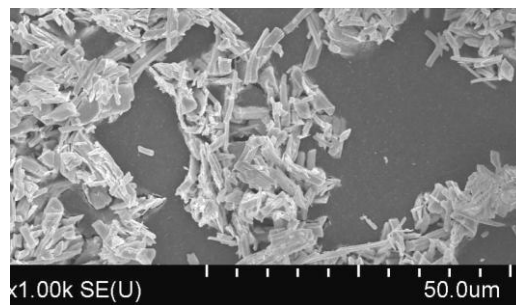
*Small agglomerations with fine particle shape*

Powder “B”



*Small agglomerations with coarse particle shape*

Powder “C”



*Large agglomerations with slender particle shape*

Fig.2a SEM images of the three different types of powder sources.

Although the characteristics of the raw powder materials are similar to each other, their sintering behavior was different to each other in terms of sintered density and microstructures along with the sintering temperatures as summarized in Fig.2b.

Powder A was fully-densified over 1700°C. Powder B was fully-densified at high temperature but densification speed was lower than powder A. Powder C has poor densification than other powders, and densification speed was the slowest.

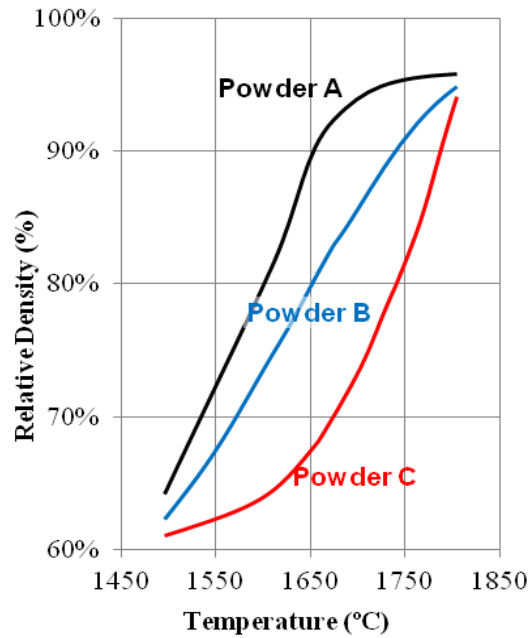


Fig.2b Sintering behavior of each powder in vacuum furnace.

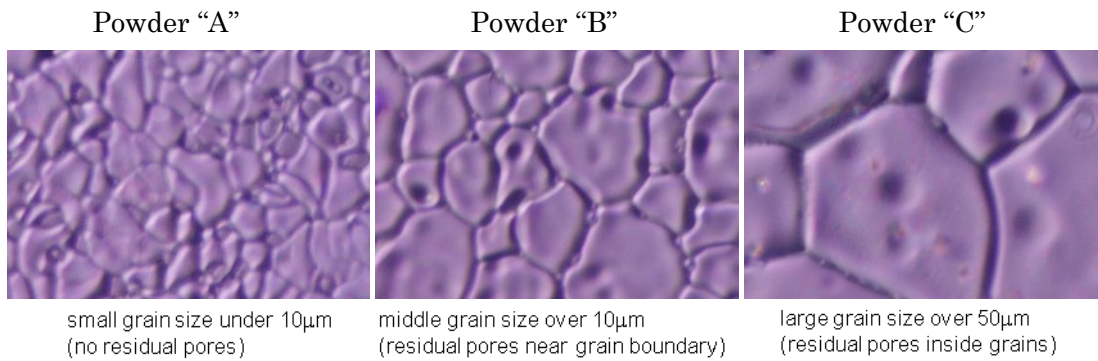


Fig.2c Microstructures by Optical Microscope (sintered at 1800°C)

Microstructure of the sintered bodies produced from each powder source was shown in Fig.2c. In powder "A" sample, grain size was smaller than 10μm but no residual pores were observed on the surface. In powder "B" sample, grain size was larger than 10μm but residual pores were observed near the grain boundary region. In the case of powder "C" sample, remarkable grain growth (grain size: >50μm) was observed and it was recognized that the residual pores were trapped inside the grains.

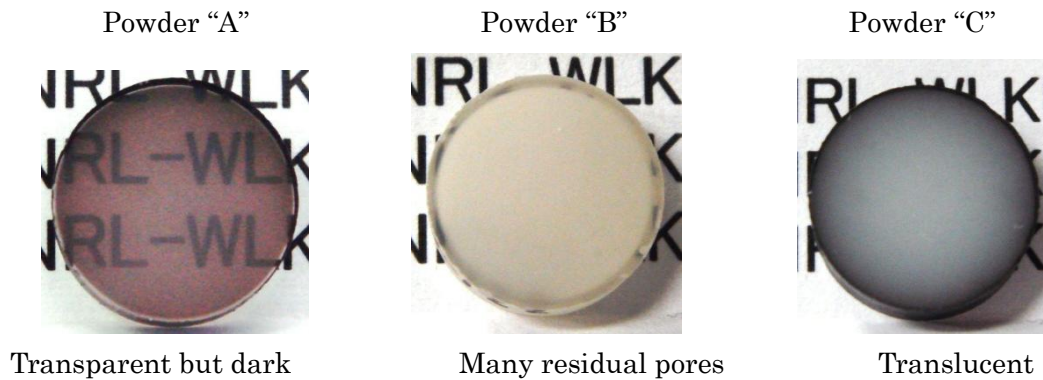


Fig.2d Appearance of 10% Yb<sup>3+</sup> doped Lu<sub>2</sub>O<sub>3</sub>

When 10%Yb doped Lu<sub>2</sub>O<sub>3</sub> ceramics were prepared by using those powders, only the powder source “A” showed transparency. But the optical quality of the prepared samples was poor without using sintering additives. Therefore, sintering additives, which do not affect laser oscillation, were introduced to produce highly transparent grade ceramics.

#### Results from 2<sup>nd</sup> and 3<sup>rd</sup> Quarter Trials

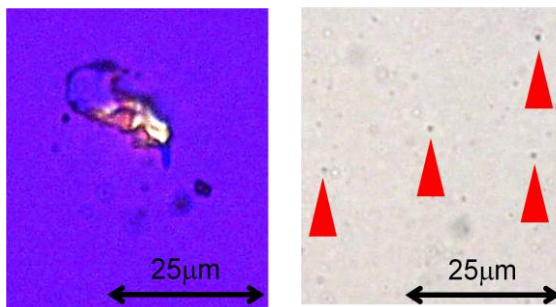


Fig. 3 Optical microscopic images of the samples done in 2011 Jan. (left) Inclusion detected by transmitted polarized microscope. (right) residual pores observed by direct transmission microscope.

As shown in the Fig.3, the microstructure of the samples prepared in 2011 Jan included many scattering sources such as inclusions and residual pores. Inclusions are typically seen when sintering aids are added, and it was mainly composed of Lu<sub>2</sub>O<sub>3</sub>, which was confirmed by elemental analysis.

Sintering aids are very effective for densification during sintering process, but if the process conditions are not optimized, the above mentioned scattering centers are generated in the materials. Therefore, it is important to control the process conditions to improve the optical quality. As a result of optimizing process conditions, the optical

transmittance of the samples prepared in 2011 April has improved in the whole wavelength ranges (please refer Fig.4 and Fig.5 for their appearance and their transmittance curves).

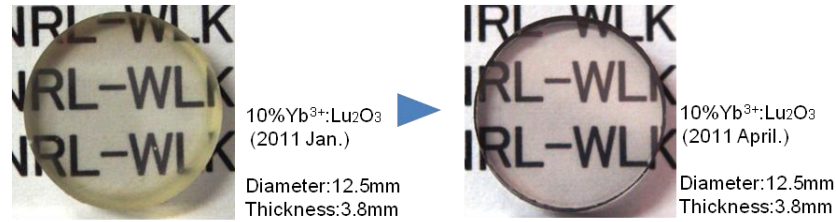


Fig. 4 Appearance of transparent  $\text{Lu}_2\text{O}_3$  ceramics in 2011 Jan. and 2011 April.

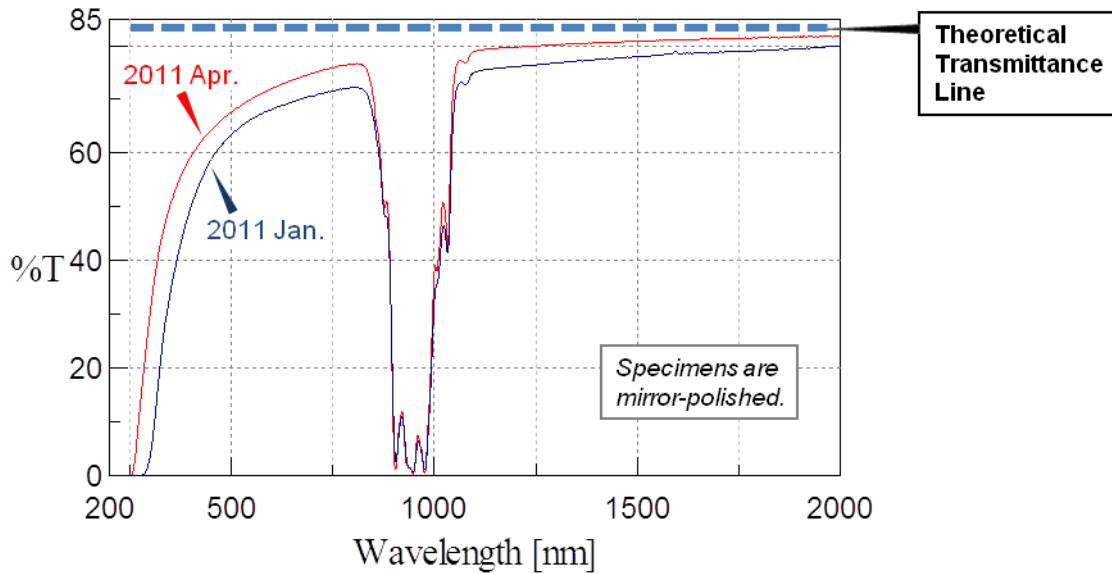


Fig.5 Comparison of transmission spectra of  $\text{Lu}_2\text{O}_3$  ceramics 2011 Jan with 2011 April.

### Further Optimization of the Fabrication Conditions in 3<sup>rd</sup> Quarter

In 2011 April, although the transmittance was improved compared to the previous samples, their optical quality is not enough. Large optical loss around the visible regions was observed. It was considered that the added sintering additives were in excess condition, and they were segregated around the grain boundary regions, causing scattering centers or lattice defects which caused Rayleigh scatterings. Therefore, we reduced the amount of sintering additives and optimized the fabrication process conditions to control those small scattering sources. Detailed fabrication flow chart is shown in Fig.6.

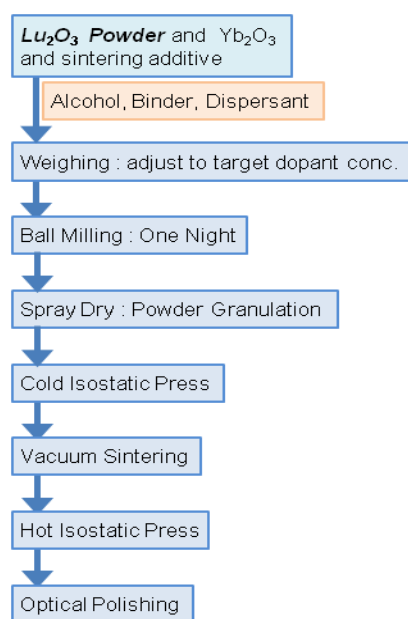


Fig. 6 Fabrication process for transparent Lu<sub>2</sub>O<sub>3</sub> ceramics.

As a result, an improvement of the transmittance in the visible wavelength ranges was achieved in 2011 June. (Please refer Fig.7 and Fig.8.)

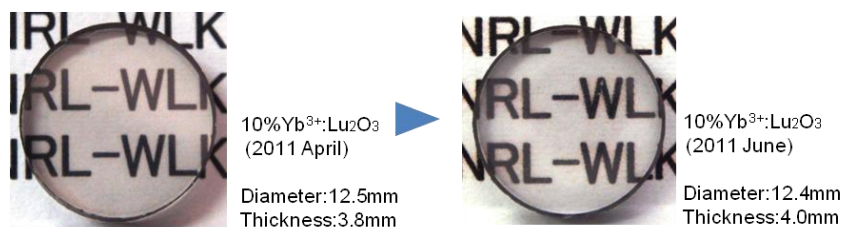


Fig. 7 Appearance of transparent Lu<sub>2</sub>O<sub>3</sub> ceramics in 2011 Apr. and 2011 Jun.

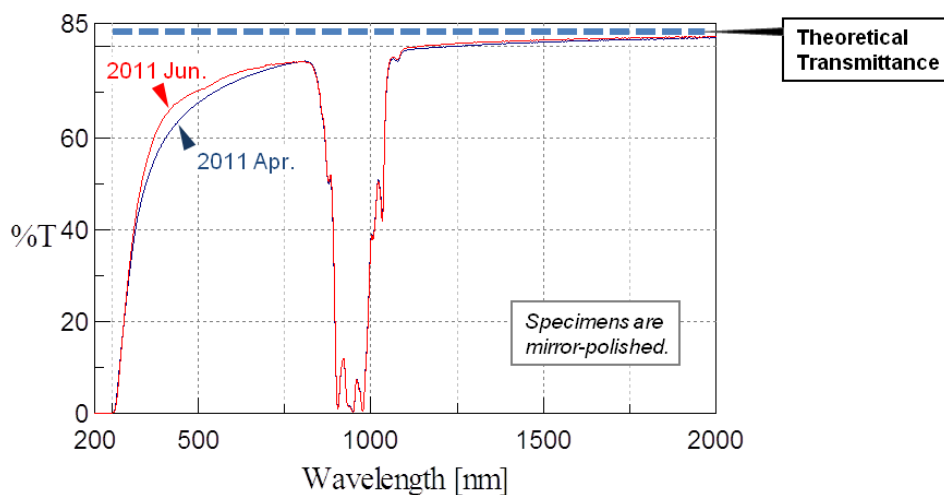


Fig.8 Comparison of transmission spectra of Lu<sub>2</sub>O<sub>3</sub> ceramics 2011 Jun. with 2011 April.

### Evaluation of Optical Quality of Final Samples for laser oscillation test

By further optimization on the whole fabrication process, we could achieve high optical quality 10%Yb<sup>3+</sup>:Lu<sub>2</sub>O<sub>3</sub> in 2011 July (see Fig.9 for its appearance). In Fig.10, it was confirmed that the optical transmittance around the infra-red regions reached very close to the theoretical transmittance. The optical losses around 940nm (excitation wavelength) and 1030nm (oscillation wavelength) are found to be very small. Although the scattering condition around the visible regions was greatly improved compared to the previous samples, small optical losses due to scattering or absorption were observed from UV to 600nm regions.

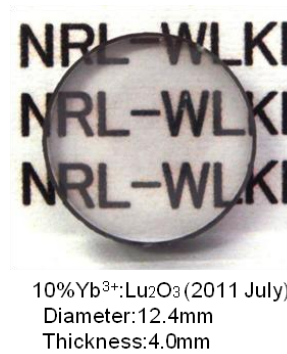


Fig. 9 Appearance of Yb doped Lu<sub>2</sub>O<sub>3</sub> ceramics

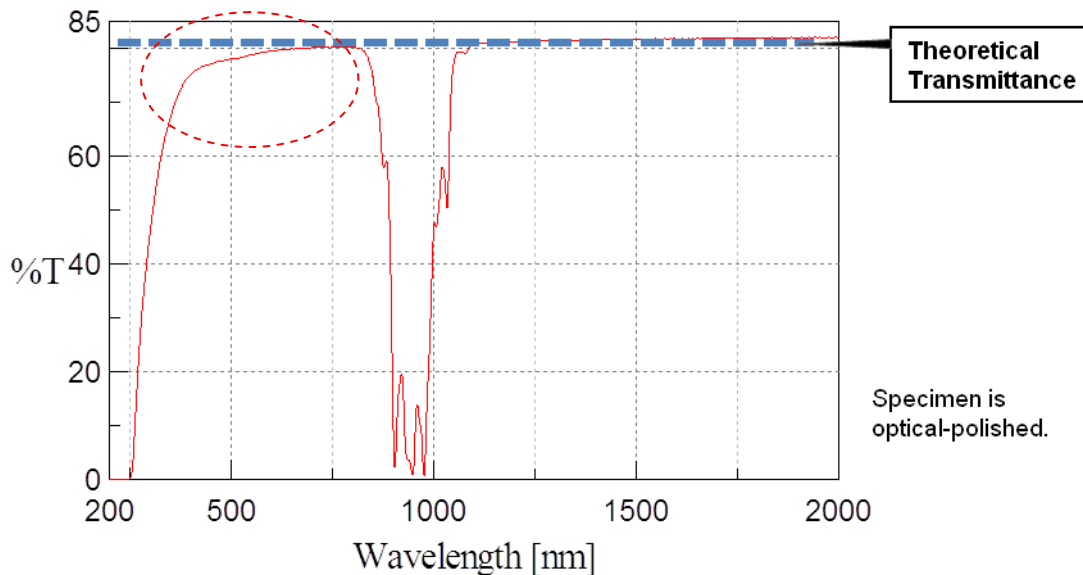


Fig. 10 Transmittance spectra of 10% Yb doped Lu<sub>2</sub>O<sub>3</sub> ceramics (2011 July)

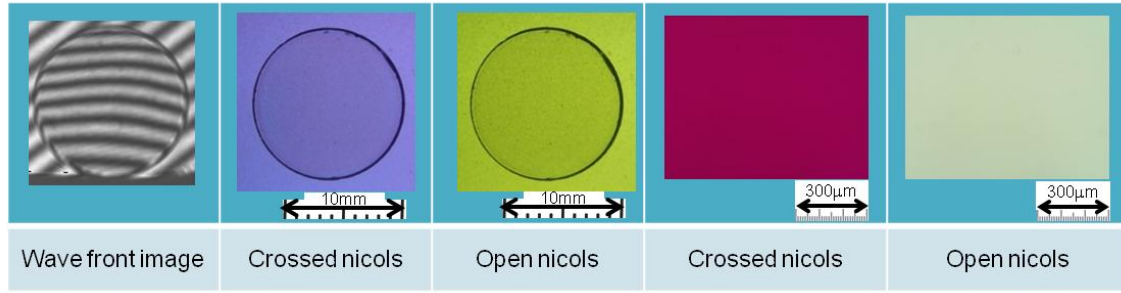


Fig. 11 Optical homogeneity for Yb doped  $\text{Lu}_2\text{O}_3$  ceramics (2011 July)

Its optical homogeneity was characterized by interferometry, and under polarizer and optical microscope in macro and micro-scale as seen in Fig.11. These results showed that the produced sample is high quality with an optical grade. Therefore, it was selected for laser experiment test.

#### **Summary of Progress of $\text{Yb}^{3+}:\text{Lu}_2\text{O}_3$ Ceramics in this Project**

The progress on the optical quality of the 10%  $\text{Yb}^{3+}$  doped  $\text{Lu}_2\text{O}_3$  ceramics are summarized in Fig.12 and 13. It is clear that the quality was improved with the timeline of the project. Although the transmittance of the sample achieved in 2011 July reached very close to the ideal line, some scattering or absorption losses around UV to visible wavelengths regions were observed. Accordingly, it can be suggested that there are still many items to investigate in order to improve the optical properties in these wavelengths.

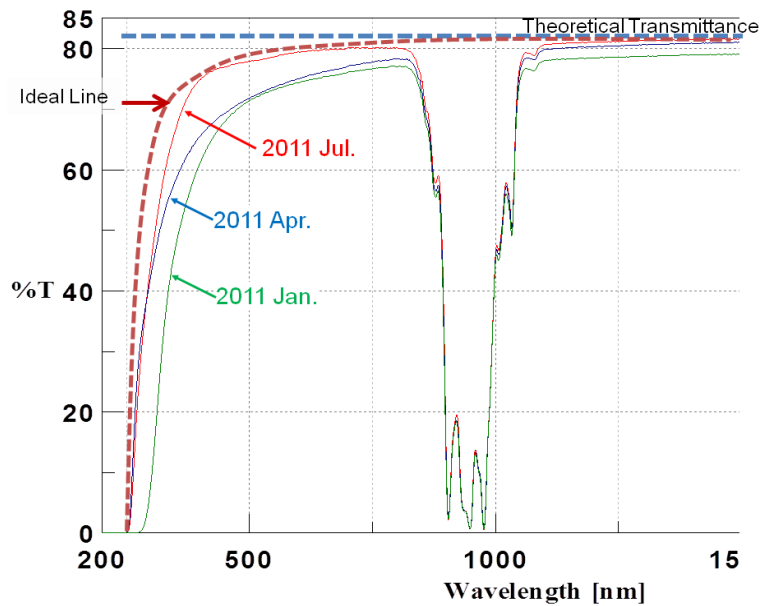
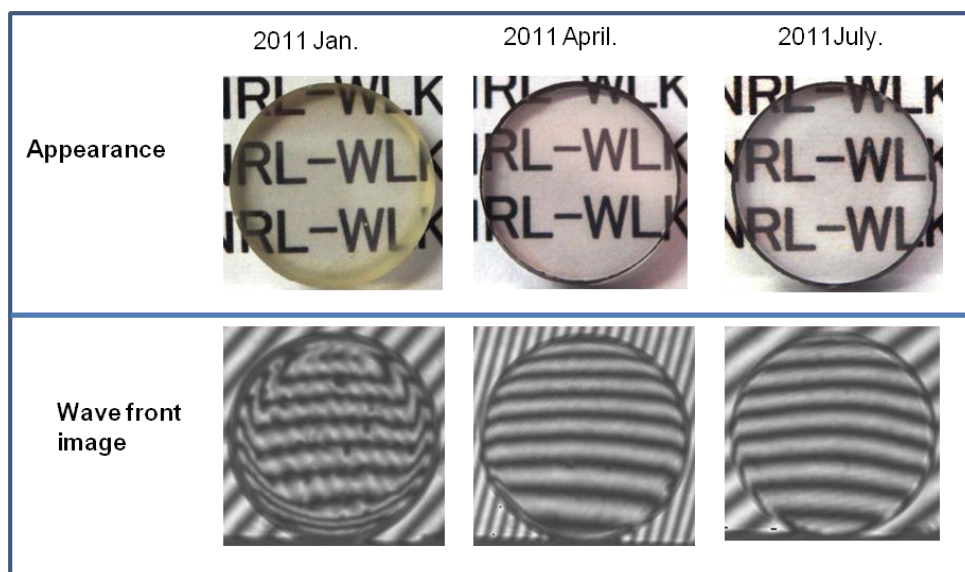


Fig. 12 Transmittance spectra of 10%  $\text{Yb}^{3+}$  doped  $\text{Lu}_2\text{O}_3$  ceramics.



*Specimens are optical-polished.*

Fig. 13 Appearance and optical homogeneity of Yb:Lu<sub>2</sub>O<sub>3</sub> ceramics.

#### Evaluation on Reproducibility of the 10%Yb:Lu<sub>2</sub>O<sub>3</sub> Ceramics

By applying the same powder source and same fabrication process, relatively high reproducibility was confirmed in terms of optical transmittance and interferometry as summarized in Fig.14.

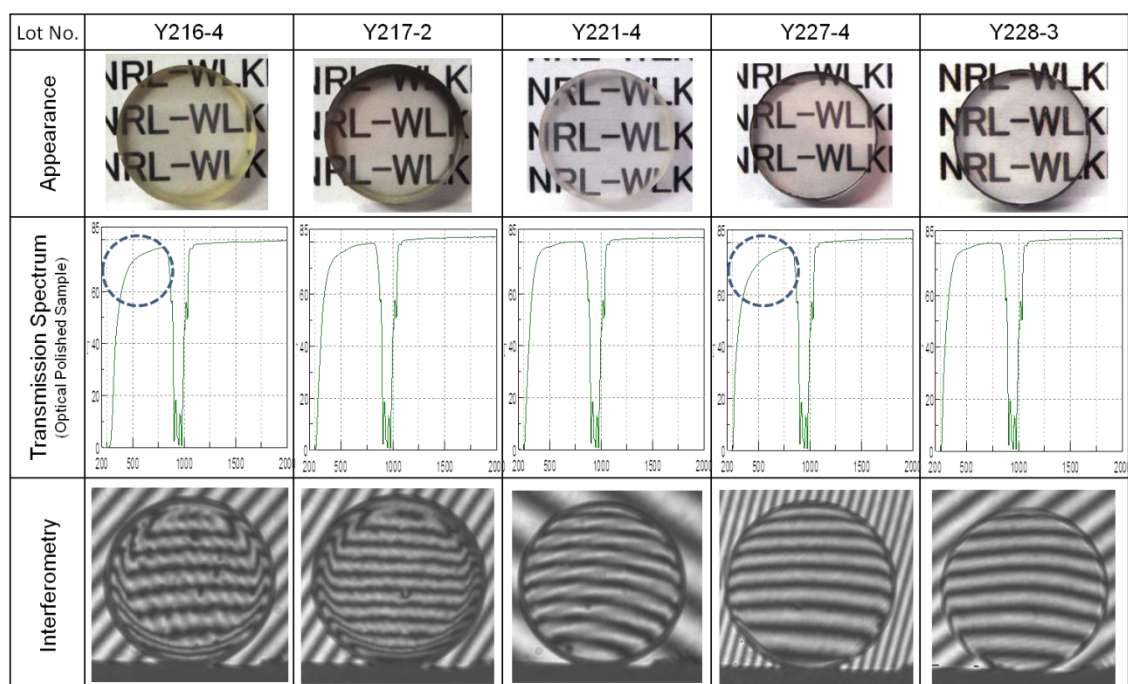


Fig. 14 Reproducibility and optical measurement for Yb:Lu<sub>2</sub>O<sub>3</sub> Ceramics of 5 batches.

### Evaluation of the samples by laser oscillation test

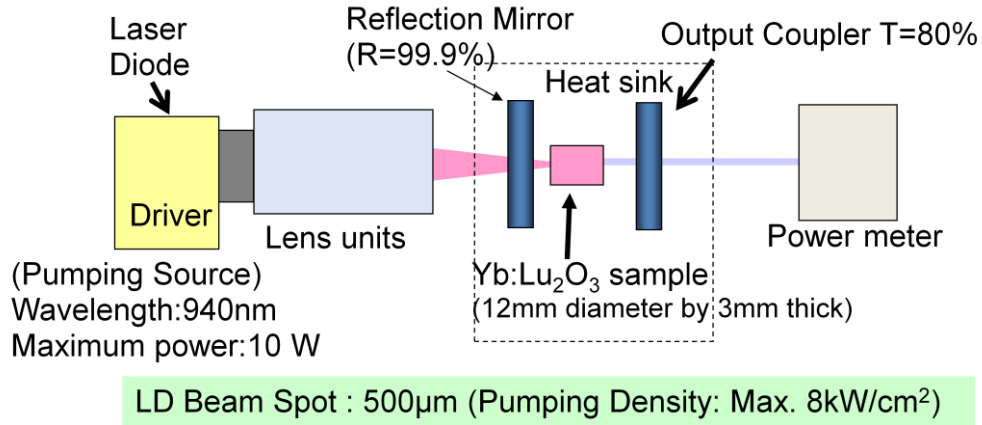


Fig. 15 Laser set-up for 10% Yb<sup>3+</sup>:Lu<sub>2</sub>O<sub>3</sub> ceramics

End-pumping system was performed in this laser experiment. 940nm LD was used as a pumping source for this laser oscillation experiment. Maximum output of the LD is ca. 10W.

Both faces of samples were polished to laser grade (flatness:  $\lambda/4$ , micro-roughness:  $<0.1\text{nm}$ ) but anti-reflection was not coated.

### Results of Laser Experiment Test

Laser oscillation results are summarized in Fig.16a. Although laser oscillation was confirmed using the 10%Yb:Lu<sub>2</sub>O<sub>3</sub> Ceramics, laser oscillation suddenly stopped at a certain input power. When the input power was further increased, damage to the samples was occurred.

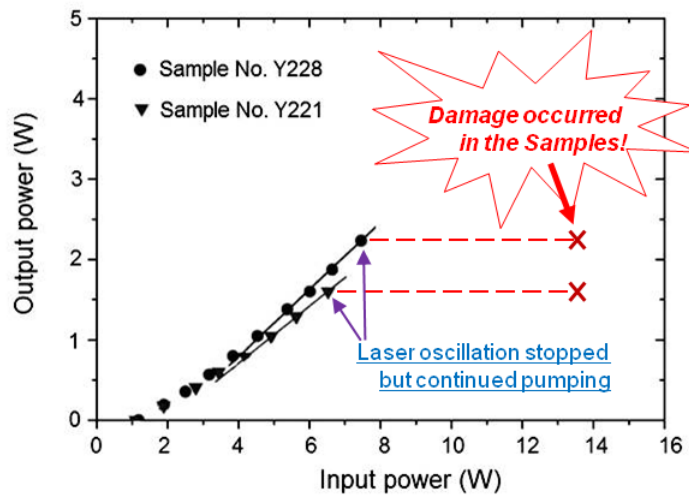


Fig.16a Laser oscillation results using the produced samples

Appearance of Yb:Lu<sub>2</sub>O<sub>3</sub> ceramics after laser experiment are shown in Fig.16b. One sample was melt-down on the surface, and another one was cracked.

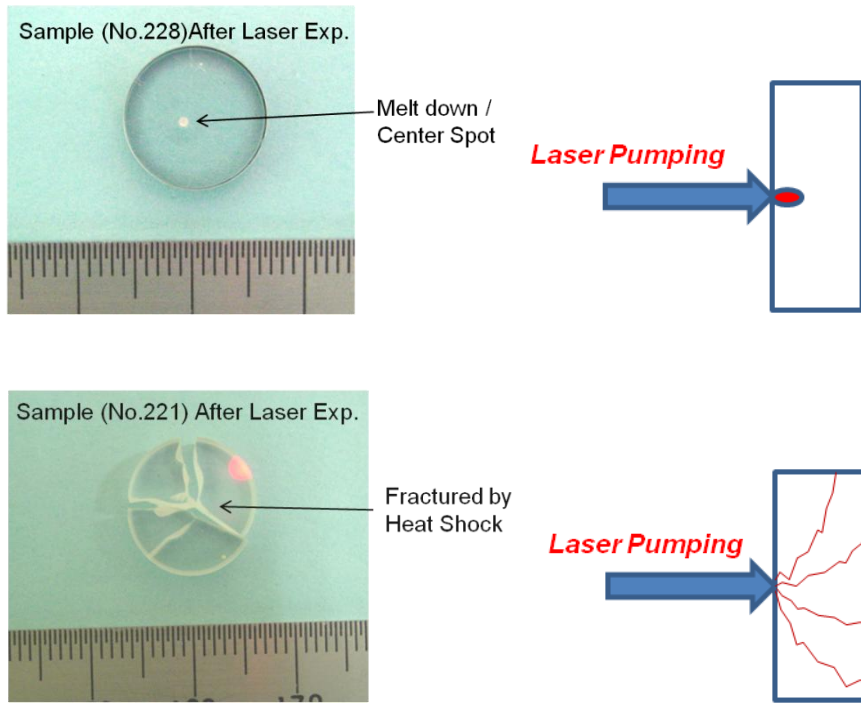


Fig.16b Appearance of Yb:Lu<sub>2</sub>O<sub>3</sub> ceramics after laser experiment.

## Conclusions

### Material Development

- 1) Succeeded in the production of highly transparent Yb:Lu<sub>2</sub>O<sub>3</sub> ceramics around near infra-red area (laser oscillation wavelength)
- 2) Scattering sources such as residual pores, grain boundary phases, birefringence were not observed in the produced samples but some inclusions were observed under optical microscope.
- 3) Internal stress was not detected by polarizer.
- 4) Slightly curved transmission wavefront images were detected by interferometry, showing that the optical homogeneity is relatively high.
- 5) Scattering or absorption around UV to visible regions was detected, but the cause of such problems needs further investigations.
- 6) High reproducibility (higher than 60%) was confirmed.

### Laser Experiment

- 1) Laser oscillation was confirmed but the oscillation suddenly stopped at a certain input power during excitation.
- 2) Cracking or meltdown problems occurred from where the laser excitation beam was focused.
- 3) The following reasons can be considered;
  - 3-1) Ceramic gain medium is too thick: Accumulated heat at the pumping area caused cracking or melting problems.
  - 3-2) Insufficient oscillation conditions: Cooling, pumping density, Fresnel scattering (non-AR coated samples)
  - 3-3) Insufficient optical quality: Scattering at visible wavelength regions induced Rayleigh scattering around excitation wavelengths and laser oscillation wavelengths (?)

Further investigations on 3-1~3-3 are necessary to be able to achieve the targeted results.